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Title: Advanced Quantum Emitters: Chemistry, Photophysics, Integration and Application

Author(s): Hollingsworth, Jennifer Ann

Intended for: Northrop Grumman/Brookhaven Lab Workshop Building Collaborations with National Labs (Limited audience)

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Advanced Quantum Emitters:

Chemistry, Photophysics, Integration and Application

Jennifer A. Hollingsworth
Materials Physics & Applications Division
Center for Integrated Nanotechnologies
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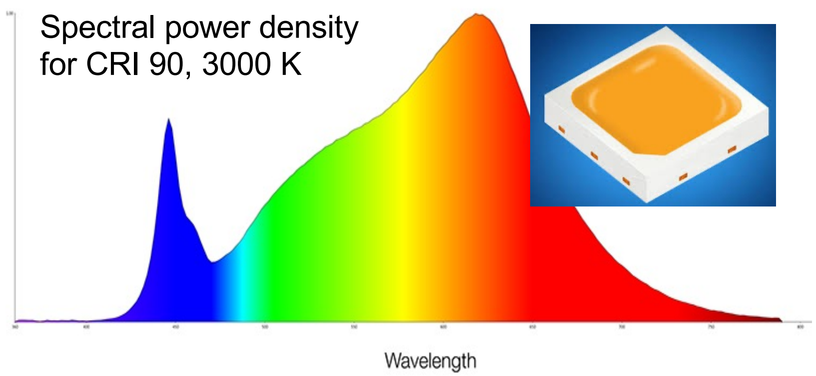
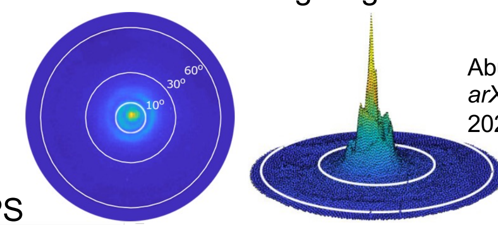
Light-Emitting Nanomaterials Needed for Demanding Applications

- **Bulk**
  - Down-conversion phosphors
  - Directly-driven sources in electroluminescent devices
- **Single Emitter**
  - Reliable single-photon sources (SPSs) for QIS

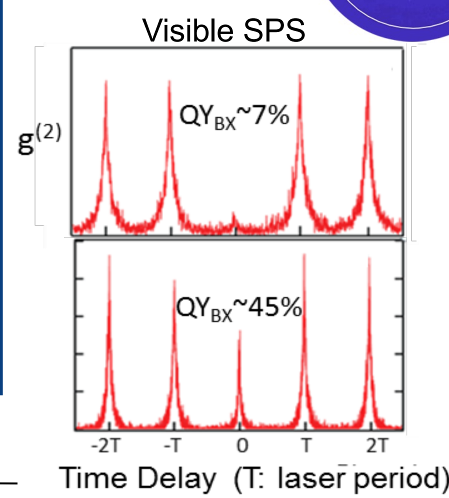


Osram's QD-enabled *Osconiq S 3030QD* mid-power LED: Color rendering + efficacy

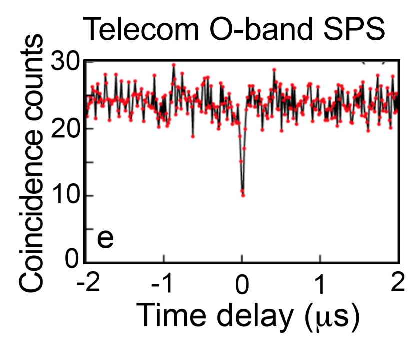
Directional free-standing single emitter



Abudayyeh et al. *arXiv:2005.11548* 2020



Mangum et al. *Nanoscale* 2014 2014, 6, 3712



Krishnamurthy et al. *ACS Nano* 2021, 15, 575

How do we ensure QDs are “alive” when we need them?

- **Bulk**
  - Osram’s QDs are encapsulated
- **Single Emitter**
  - Core/shell heterostructuring suppresses processes that turn QDs “off”

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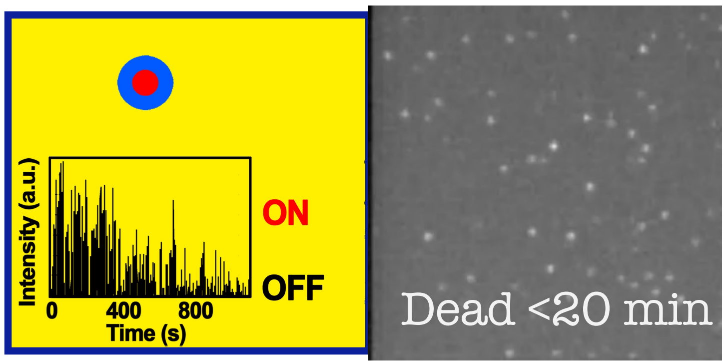
Detecting emission from single QDs in the solid-state

- **Wide-field optical microscopy**
  - *Core-only QD*: Polymer coating is needed
  - *Core/thin-shell*: Imaged without protective coating, but blinks/bleaches
  - *Core/thick-shell*: Blinking and bleaching suppressed

CdSe/thin-CdS



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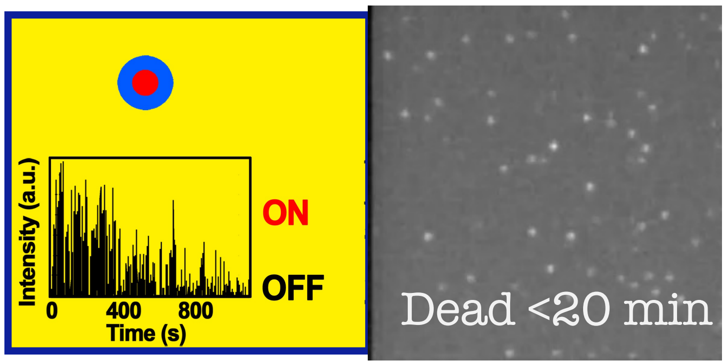
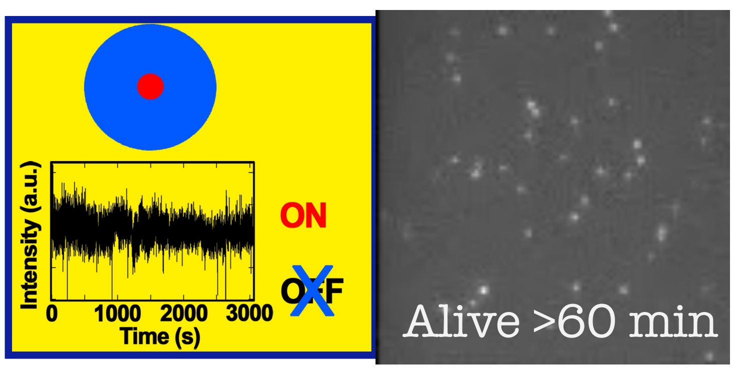
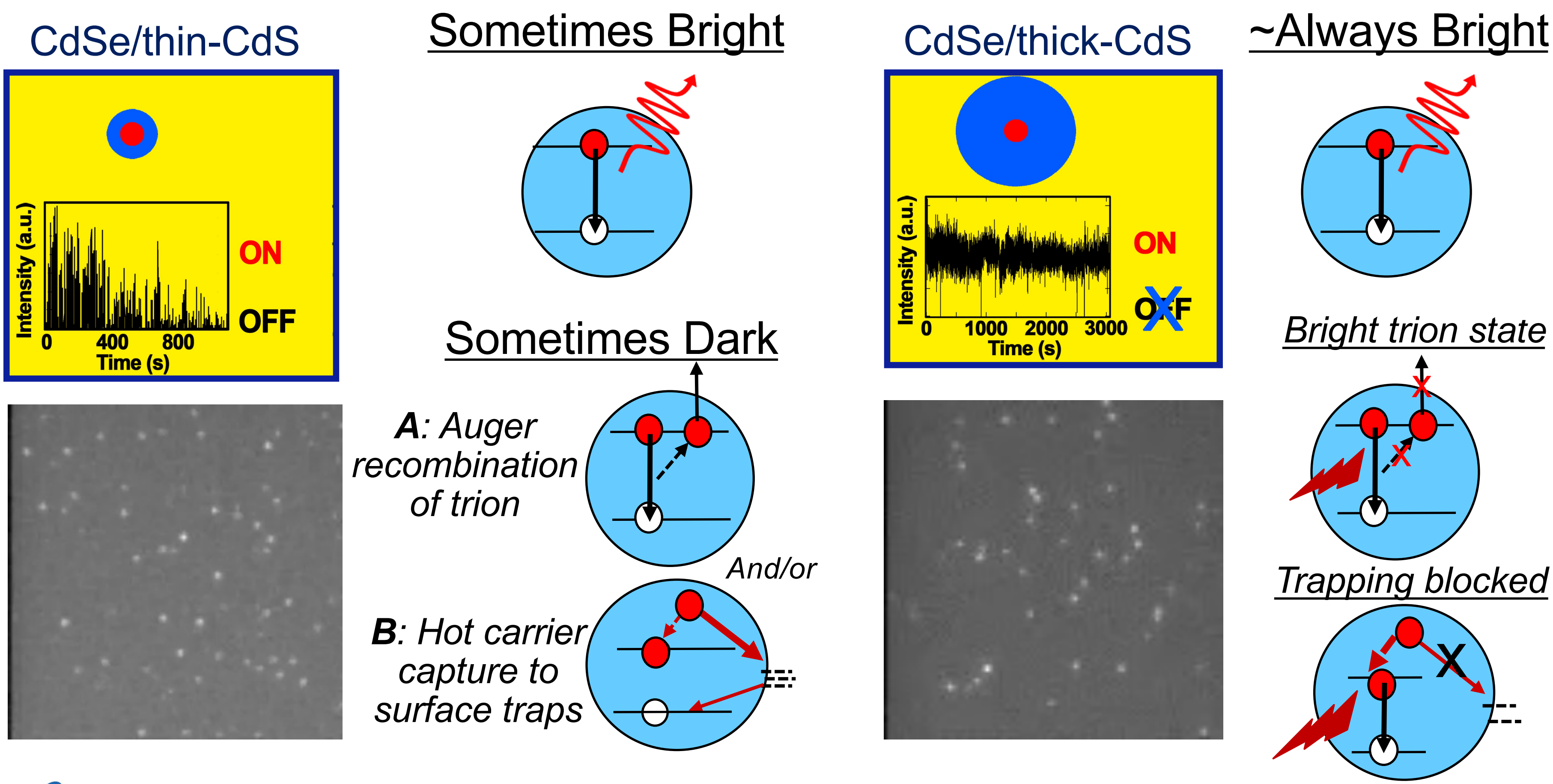


CdSe/thick-CdS
"Giant" QDs (gQDs)



Since Chen *et al. J. Am. Chem. Soc.* **2008**, 5026

The gQD's thick shell impacts blinking in two ways



Two blinking mechanisms: Galland et al. *Nature* **2011**, *479*, 203

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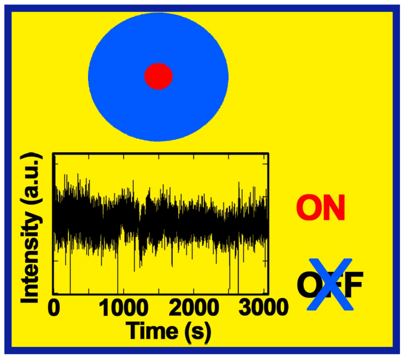
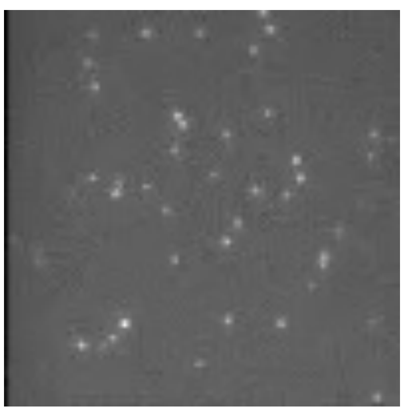
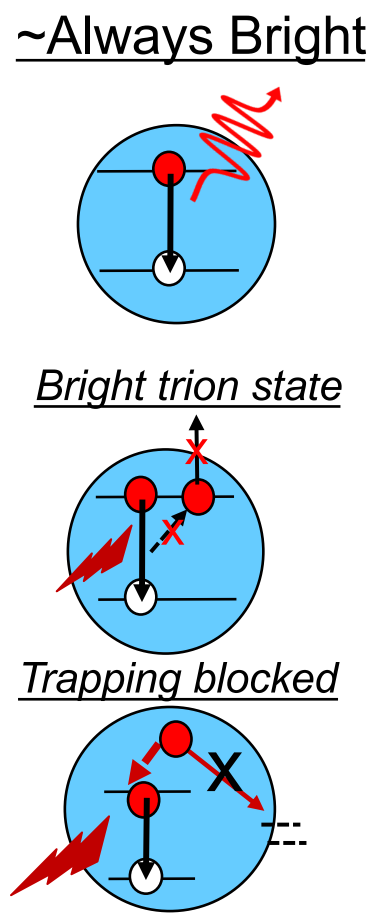
CdSe/thick-CdS



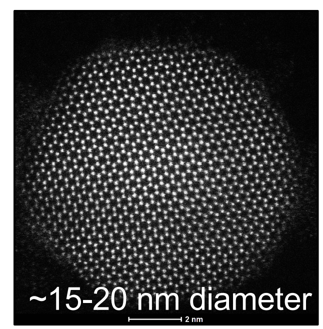




Key design parameters

Suppressing hot-carrier trapping
- ✓ Core isolation

Suppressing Auger-mediated blinking
- ✓ Large volume
- ✓ Type II band structure
- ✓ Possibly an alloyed interface (potential energy smoothing



Rational heterostructuring: Hollingsworth *Chem. Mater.* **2013**, *25*, 1318

gQD properties now span large spectral range

- **Family of Giant QDs**

CdSe/CdS:

Chen *et al. J. Am. Chem. Soc.* **2008**, 5026

Ghosh et al. *J. Am. Chem. Soc.* **2012**, 9634

InP/CdS & *InP/CdSe:*

Dennis et al. *Nano Lett.* **2012**, 5545

Buck et al. *Adv. Funct. Mater.* **2019**,1809111

PbSe/CdS & *PbS/CdS*

Hanson et al. *J. Am. Chem. Soc.* **2017**, 11081

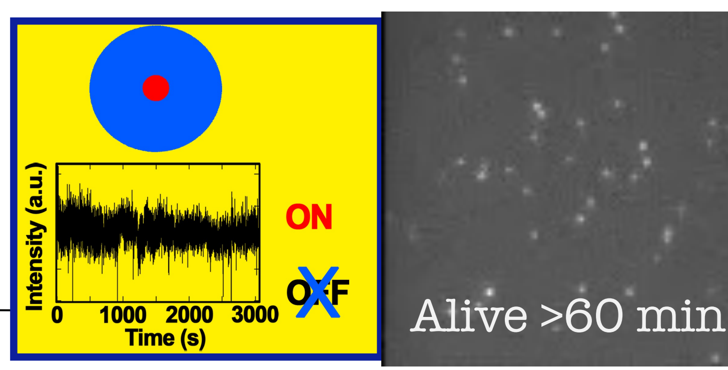
Krishnamurthy et al. *ACS Nano* **2021**, 575

CdSe/CdS "giant tetrapods"

Mishra et al. *Nature Commun.* **2017**, 15083

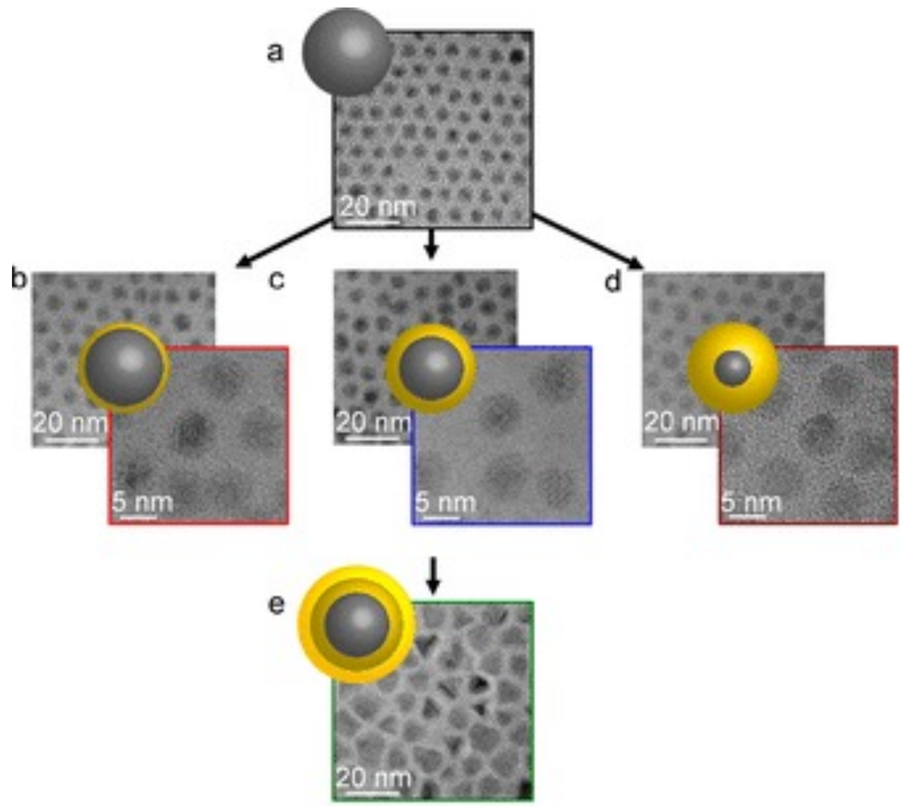
CdZnSSe/ZnS:

McBride et al. *J. Chem. Phys.* **2020**,124713



PbS/CdS gQDs: Room-Temperature Single-Photon Source for the Telecom O and S Bands

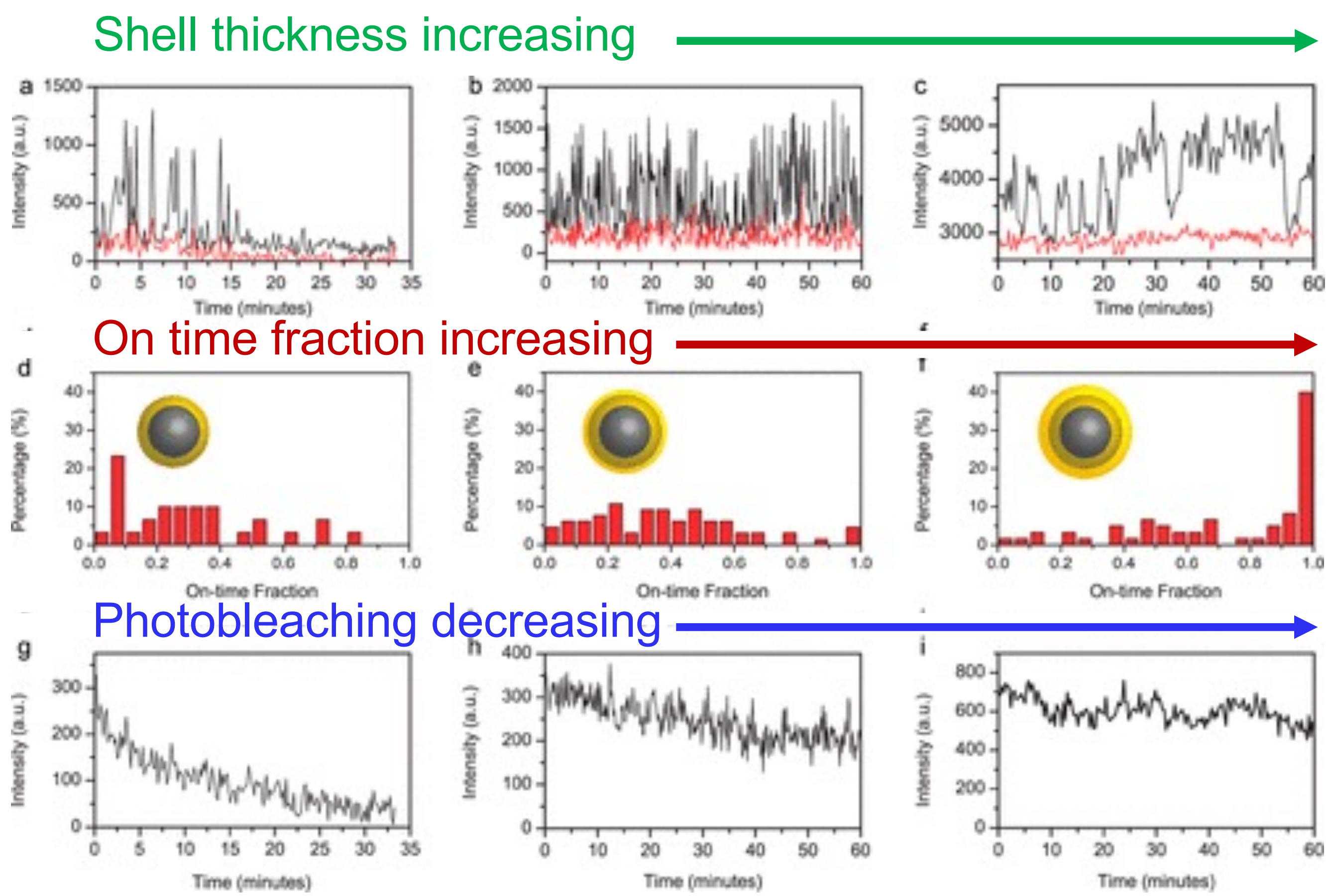
- **Using two types of shell-growth methods, we prepared a core-size/shell-thickness series for understanding structure:function correlations**



ACS Nano 2021, 15, 1, 575–587

PbS/CdS gQDs: Room-Temperature Single-Photon Source for the Telecom O and S Bands

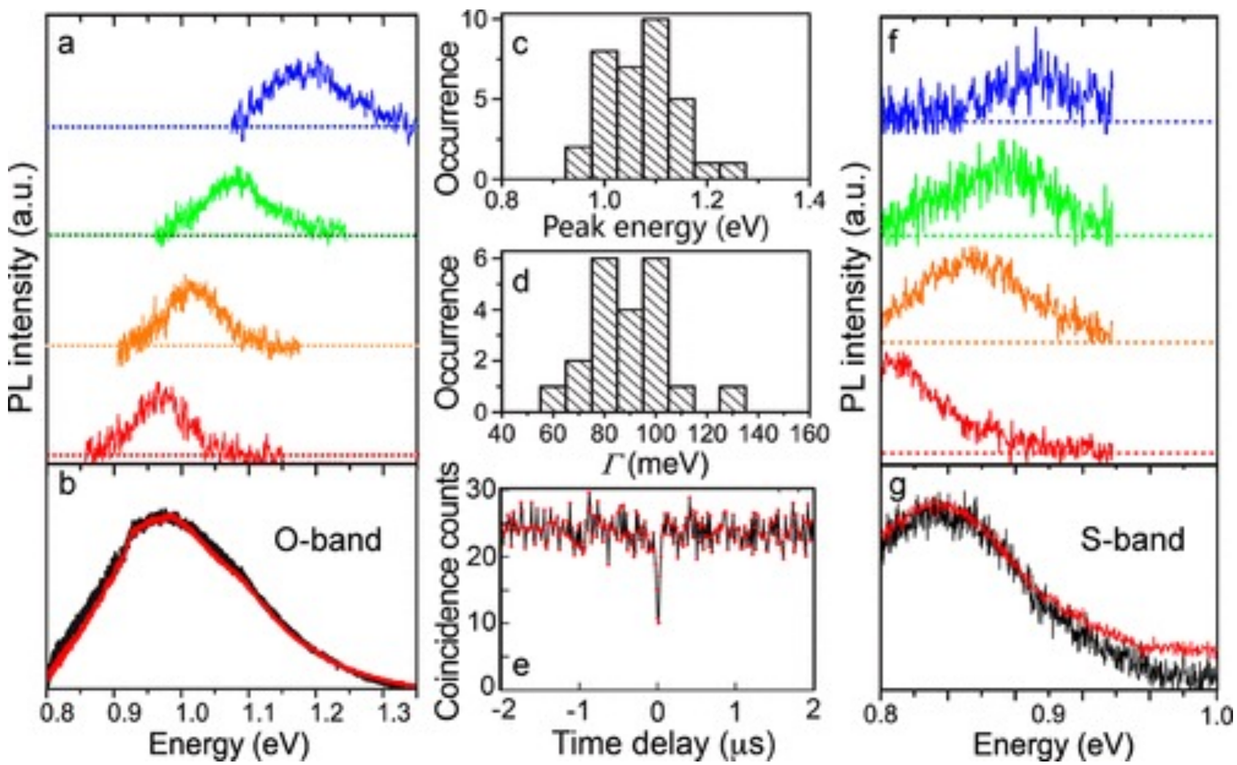
- …And, to gain control over blinking & photobleaching behavior



ACS Nano 2021, 15, 1, 575–587

PbS/CdS gQDs: Room-Temperature Single-Photon Source for the Telecom O and S Bands

- …As well as to conduct single-QD spectroscopy using standard detection methods



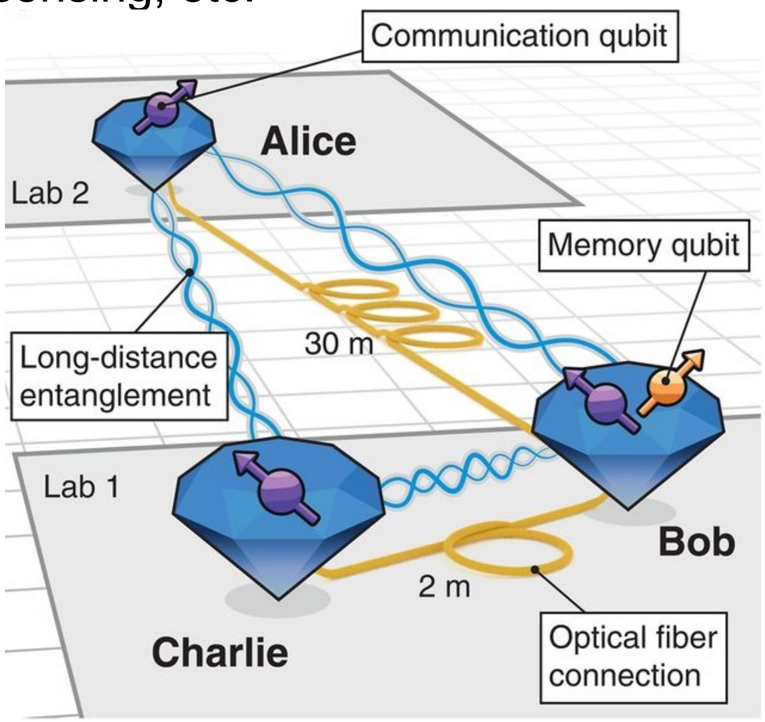
- Hallmark of a single-photon emitter: Photon-antibunching dip with a $g^{(2)}(0)$ value of ~0.4
- Shell thickness-dependent homogeneous broadening
- Greater phonon-assisted dephasing for IV–VI compared to II–VI QDs

Rm T: ACS Nano 2021, 15, 1, 575–587

Low-T Nano Lett. 2019, 19, 12, 8519–8525

Single-Photon Source (SPS) Application: Photonics-Enabled Quantum Network

- **3 Quantum devices connected via entanglement**
  - Diamond NV-center spin qubits comprise the 3 devices
  - Photons traveling through an optical fiber permit the entanglement
  - Toward secure communication, distributed quantum computing, enhanced sensing, etc.



From *Science 2021* 372, 259-264

- NV center provides optical interface, but limited to visible-light emission
- *Other single-photon sources needed to access telecom wavelengths*

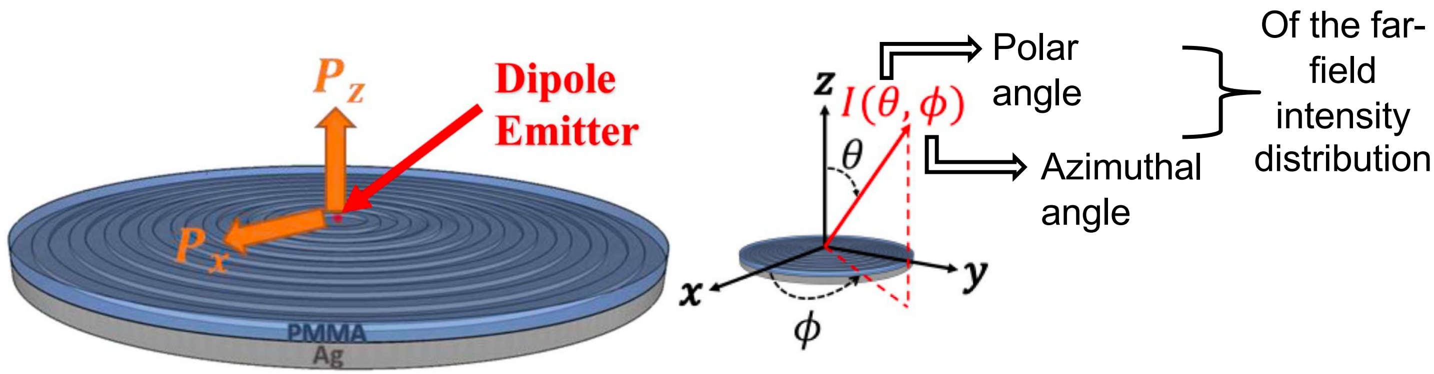
Challenge For SPS Application

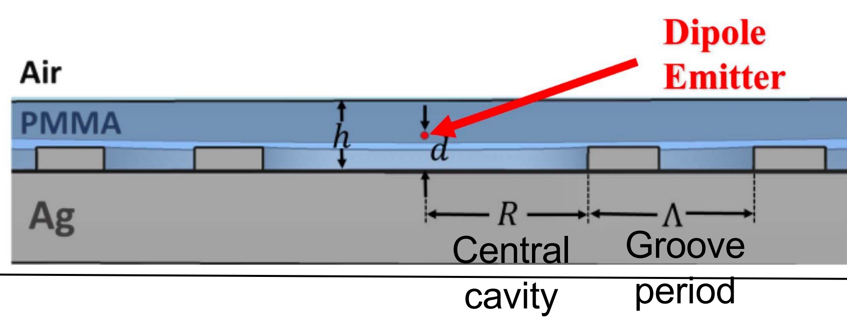
- **Free-standing SPS characterized by *omnidirectional emission***

Approach for Directional SPS: Couple Emitter to Nanoantenna

- **Unique benefits of metal-dielectric circular bullseye structures**
  - Photon source at center emits into a dielectric layer
  - Dielectric = slab waveguide → Guides light radially toward circular gratings
  - In the far-field, the interference between various diffracted waves occurs only at low angles → **Highly directional photon stream**

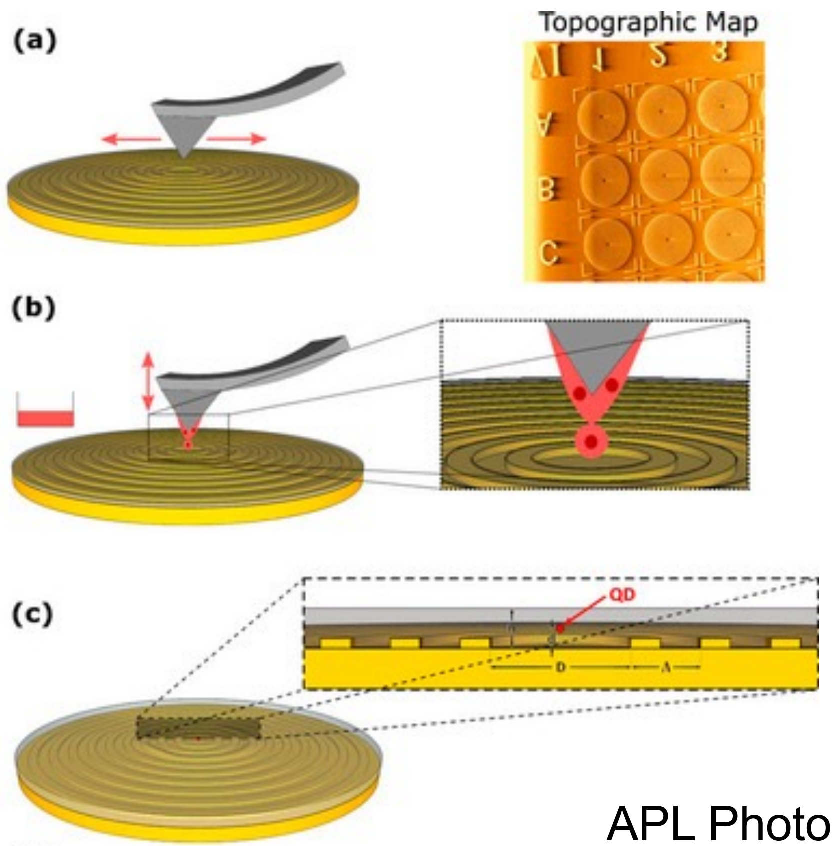
See original development by R. Rapaport & H. Abudayyeh in Quantum Sci. Technol. 2017, 2, 034004





Approach for Integrating gQD into Nanoantennas: Dip-pen Nanolithography (DPN)

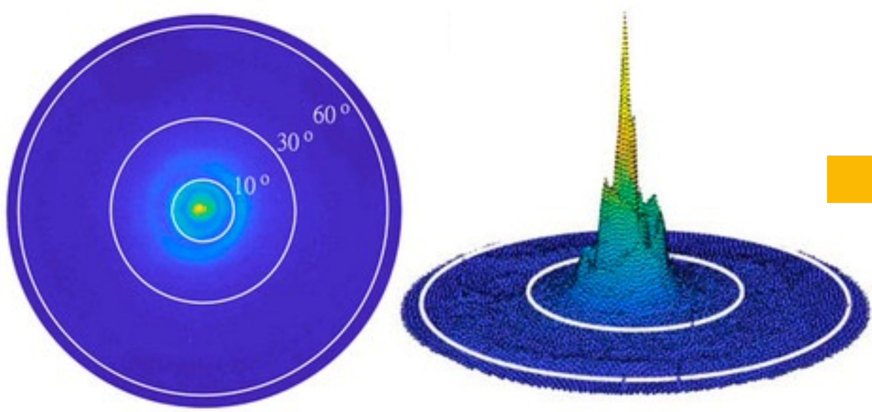
- **Unique benefits of DPN for creating functional hybrid material**
  - Ability to precisely locate a single QD in the bullseye center

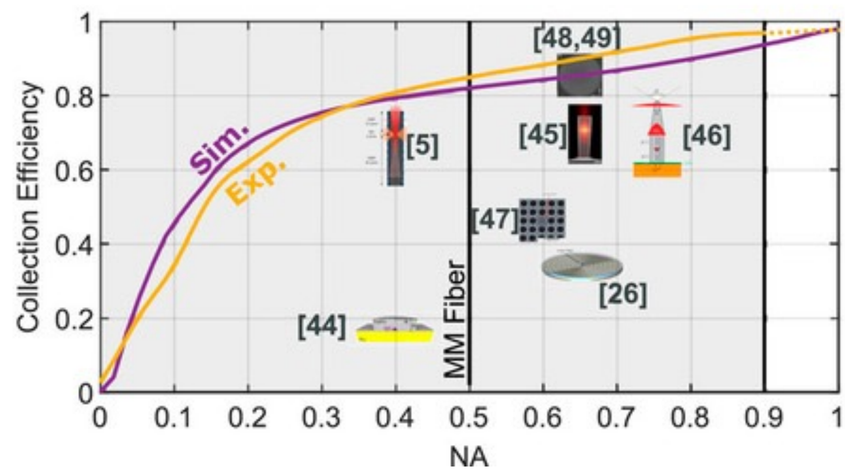


APL Photonics **2021**, 036109

Pairing Unique Quantum Emitters with Optimal Nanoantennas *Via* Deterministic Nano-integration → *New Functionality*

- **Back focal plane images of a single-photon device**
  - Record-high collection efficiency: 85% of single photons into low NA of 0.5
  - Amenable to optical fiber coupling



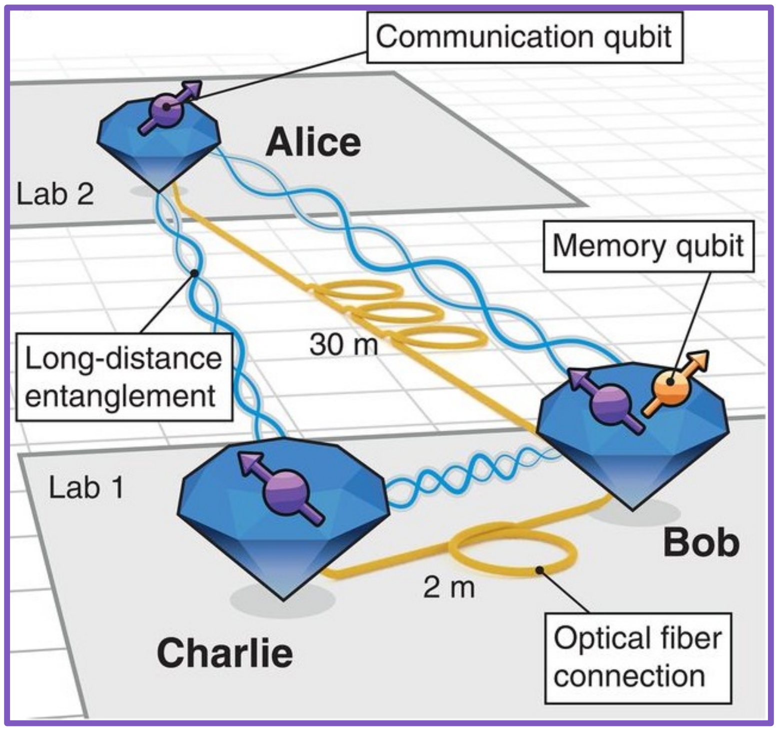


Vs. simulation & reported efficiencies for: micropillar cavities; tapered nanowires, photonic crystal cavities, other bullseye antennas

APL Photonics **2021**, 036109

Next Steps

- **Envision a quantum network with qubit and optical transduction functionalities enabled by "all-synthesized" hybrid materials**



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  Yagna Ghosh, Allison Dennis,
  Buck, Javier Vela, Yongfen Chen,
  Christina Hanson, Sachi Krishnamurthy,
  Ajay Singh, Katya Dolgopolova
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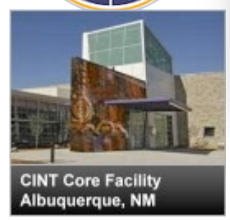
  Anastasia Blake
- Nanoantennas & other fab:
  R. Rapaport, H. Abudayyeh (Hebrew Un.); A. Malko,
  R. Bose (UT Dallas), C. Ocampo, J. Hu (MIT)



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